Digital Autoradiography technique an efficient tool for sampling procedure

P. Fichet, R. Haudebourg, C. Mougel

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DIGITAL AUTORADIOGRAPHY TECHNIQUE AN EFFICIENT TOOL FOR SAMPLING PROCEDURE

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Overview

- Context and LASE laboratory : Analytical Support to Facilities Laboratory
- Digital Autoradiography Technique
- Digital Autoradiography in support of sampling processes
- Digital Autoradiography: improvement of the selectivity
- Conclusions

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CONTEXT: MANAGEMENT OF NUCLEAR WASTES

↳ **Who?** The National Radioactive Waste Management Agency **ANDRA** is in charge of the long-term management of radioactive wastes in France

⇨ Classification of radioactive wastes as a function of their management

<table>
<tr><th>Activity - Half-life</th><th>Very short-half-life
< 100 days</th><th>Short half-life
≤ 31 years</th><th>Long half-life
> 31 years</th></tr>
<tr><td>Very low level waste</td><td rowspan="4">Management by radioactive decay</td><td colspan="2">Surface disposal facility
(CSTFA Aube facility)</td></tr>
<tr><td>Low level waste</td><td rowspan="2">Surface disposal facility (CSFMA Aube facility)</td><td>Near-surface disposal facility being studied</td></tr>
<tr><td>Intermediate level waste</td><td>Deep disposal facility at 500 meters being studied</td></tr>
<tr><td>High level waste</td><td colspan="2">Deep disposal facility at 500 meters being studied</td></tr>
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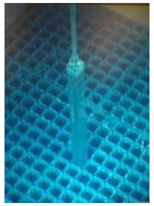
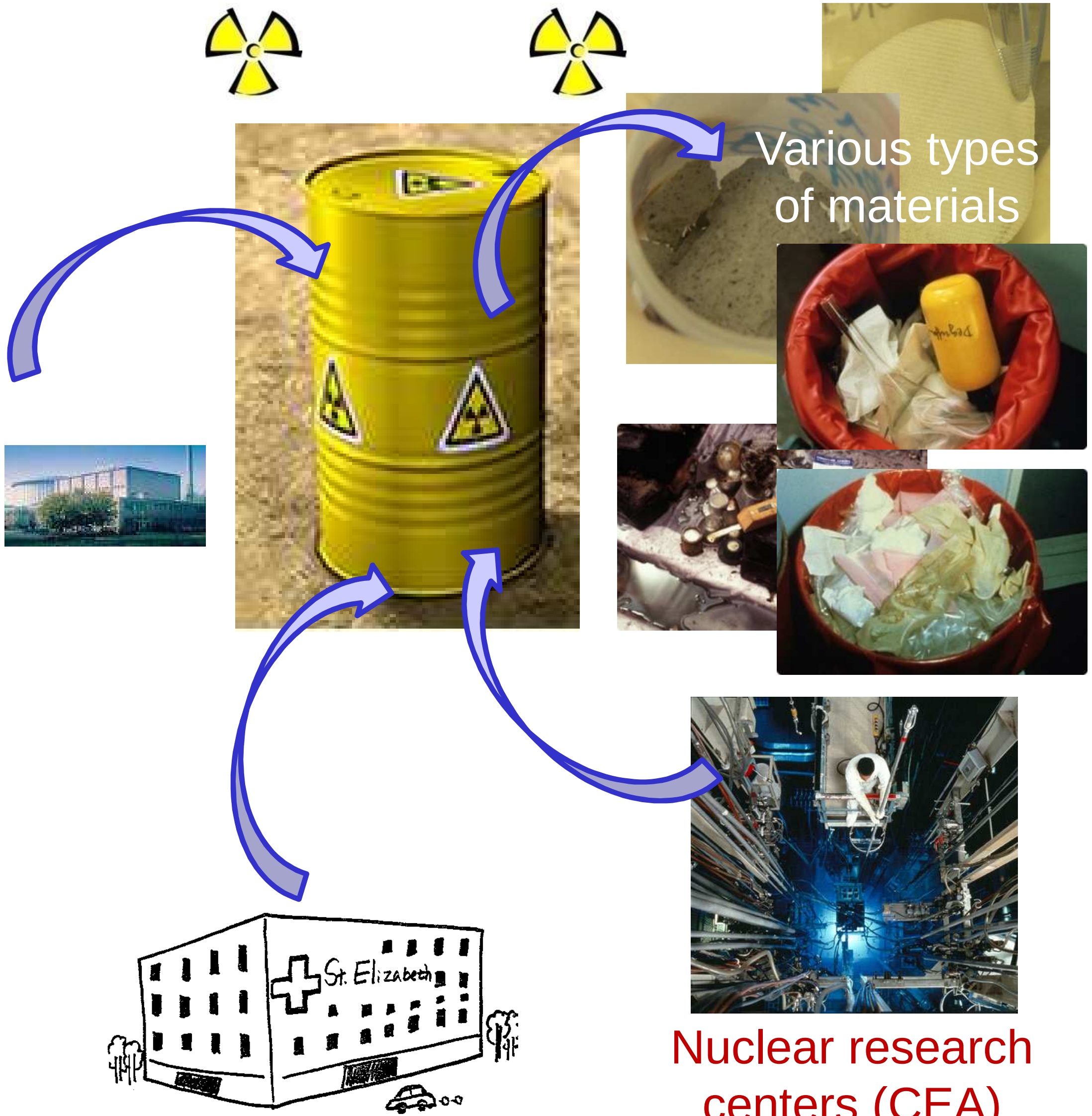
CONTEXT: ORIGIN OF LOW AND INTERMEDIATE LEVEL NUCLEAR WASTES

Nuclear power industry (EDF, AREVA)



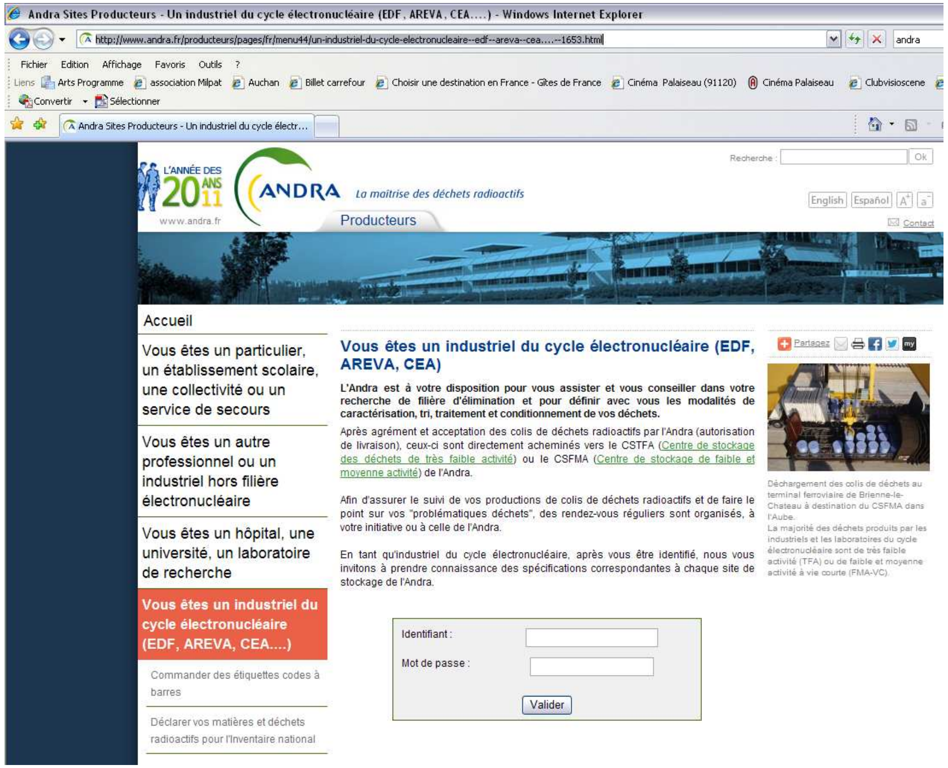


Hospitals

Nuclear research centers (CEA)

NUCLEAR WASTES ANALYSIS REQUIRED BY AUTHORITIES

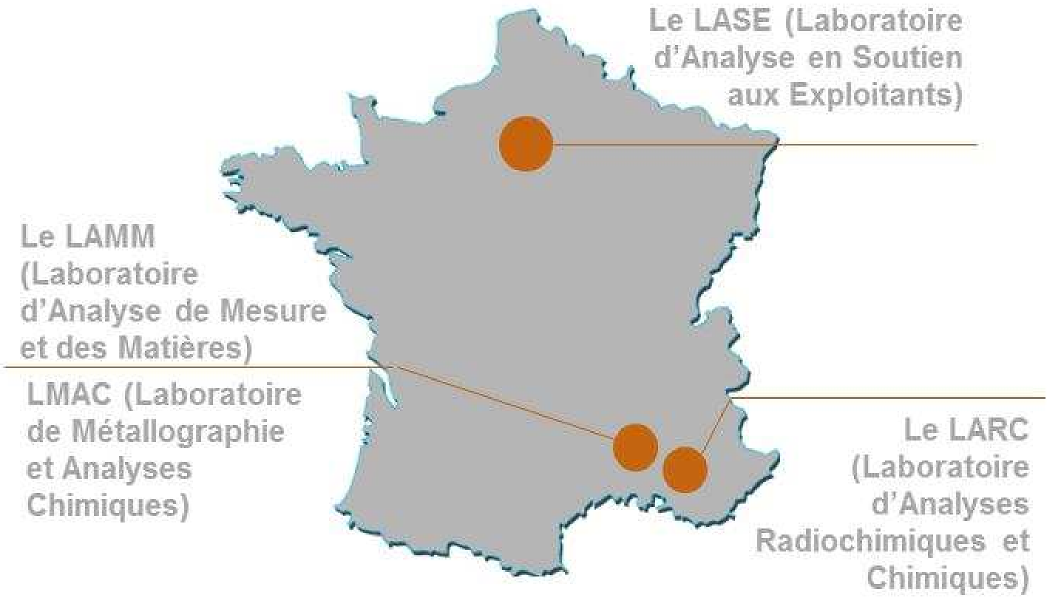
↳**Why? ANDRA** requests a characterization of nuclear wastes and specifies acceptance criteria for packages that waste producers have to respect

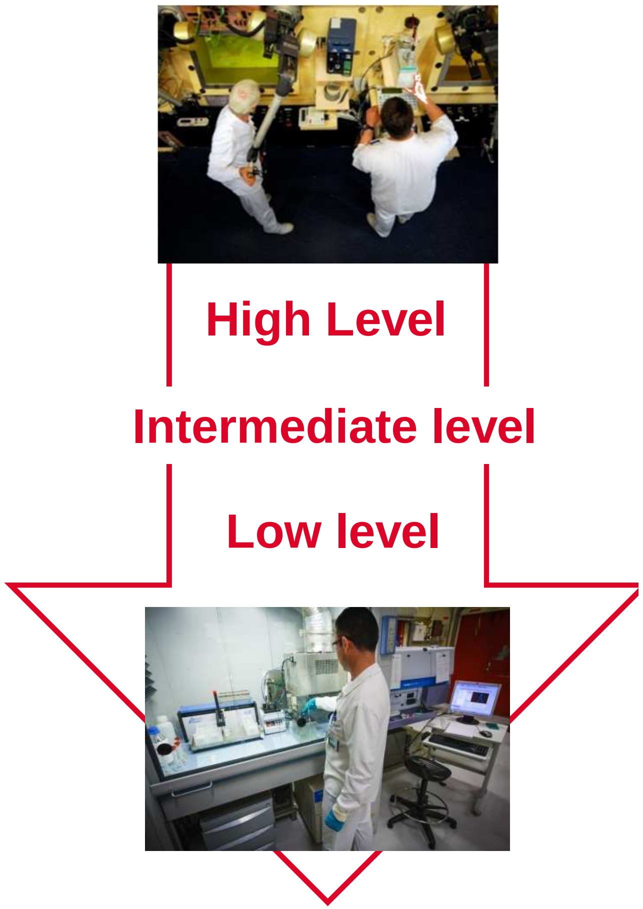


↳**Why? Characterization is one of the essential step in decommissioning projects**

ANALYTICAL CAPABILITY OF NETLAB

NucLab associated to NetLab = A CEA network of analytics and experimental tools (laboratories located on CEA sites) to assist nuclear operators during characterization programs associated to dismantling process.





LASE LABORATORY

- Different characterization techniques for low and intermediate level wastes.
- Destructive analysis (sample = 1 g)
- Radiochemistry
- Alpha, Gamma, LSC
- Elemental analysis

- In situ technique: Autoradiography

LASE: ANALYTICAL SUPPORT TO FACILITIES LABORATORY FRANCE

- ANALYSES ARE REQUIRED TO ASSESS THE SAFETY OF THE REPOSITORY SITES.

- LOTS OF MATRICES TO BE CONSIDERED, **Validation required**

Elemental analysis

23 fume cupboards
Radiochemistry

Nuclear analyses

CHARACTERIZATION OF LOW AND INTERMEDIATE LEVEL NUCLEAR WASTES

⇨ Mission of Analytical support to facilities Laboratory at CEA-Saclay

ANALYSIS OF ELEMENTS (RADIOACTIVE OR NOT) PRESENT AT TRACE LEVEL IN VARIOUS MATRICES

Chemical and radiological characterization of radioactive materials

Toxic elements, organic ligands, TOC, anions, cations

Radionuclides determined after radiochemistry (A~0.1Bq.g^{-1})

^{3}H, ^{14}C, ^{36}Cl, ^{55}Fe, ^{59}Ni, ^{63}Ni, ^{90}Sr, ^{93}Mo, ^{93}Zr, ^{93m}Nb, ^{94}Nb, ^{108m}Ag, ^{121m}Sn, ^{129}I, ^{151}Sm, ^{241}Pu, $^{238\ et\ 240}$Pu, ^{239}Pu, ^{241}Am, ^{243}Am, ^{232}U, ^{234}U, ^{235}U, ^{238}U

Wipes, technological wastes (tissues, gloves), concretes, ion exchange resins embedded in organic polymers, metals, muds, sludges, oils…

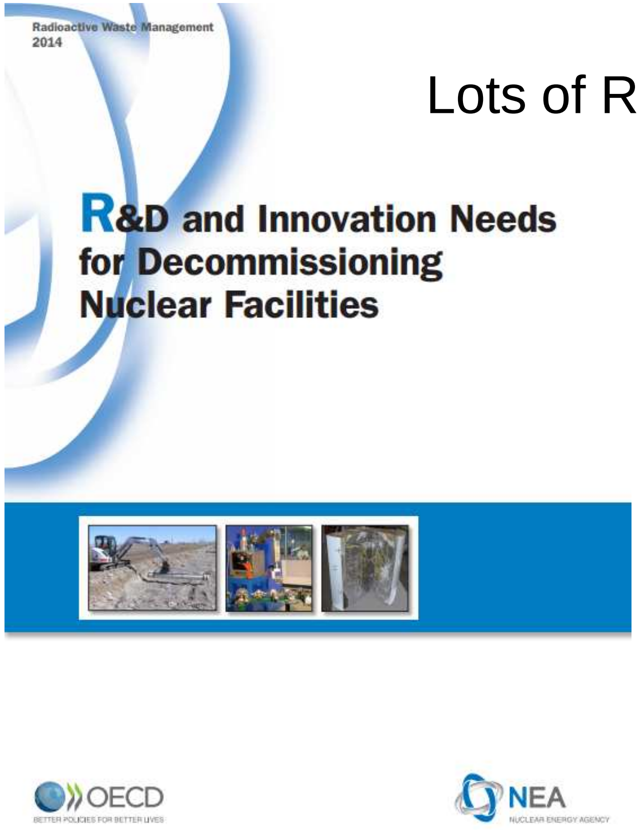
⇨ Main Radionuclides that must be investigated in priority

- Easy to measure:
  - gamma emitters
- Difficult to measure
  - Alpha emitters
  - Beta emitters
    - H-3
    - C-14
    - Cl-36
    - Sr-90

CHARACTERIZATION IS VERY IMPORTANT FOR D&D PROJECTS

Destructive techniques need less than 1 g of sample for digestion process



Lots of Radwastes must be characterized

Innovation is required for in situ techniques, for techniques allowing a better sampling process

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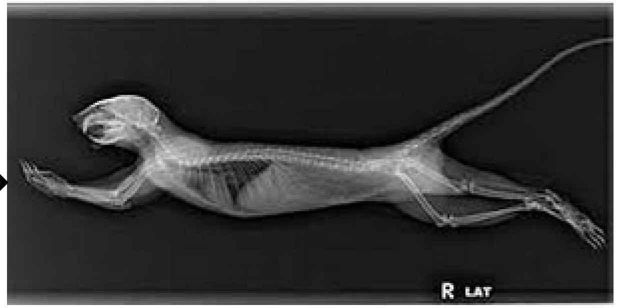
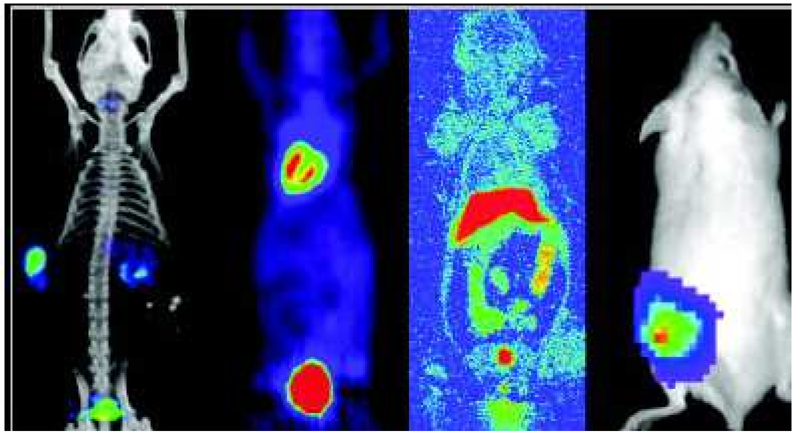
Radiography

Source



Detector

Autoradiography



Detector

Interests of biologists: H, C, S, … radionuclides difficult to measure

Digital Autoradiography

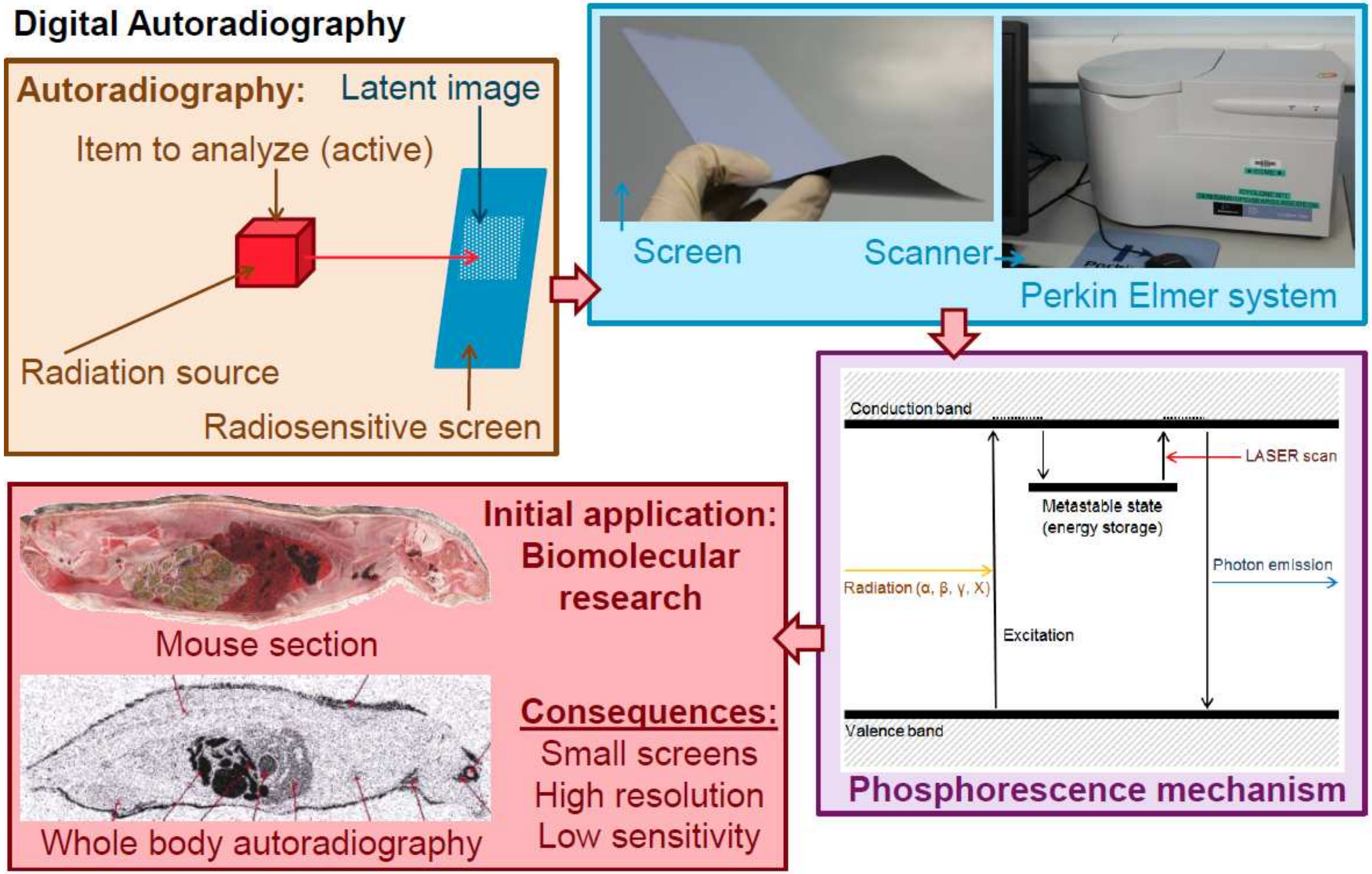
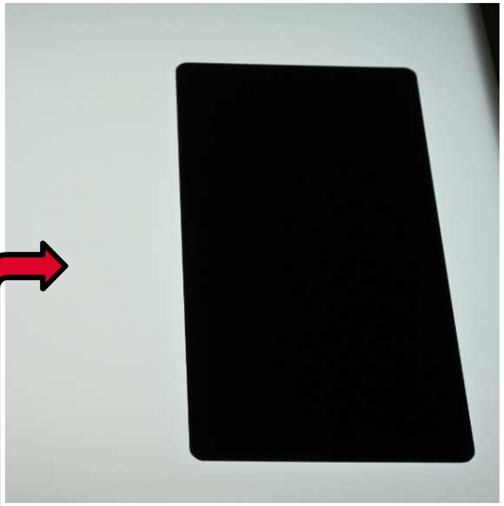


Image of radioactivity = efficient tool for sampling procedure

DA TECHNIQUE

Screens can be reused hundreds of times

(1) Screen initialization

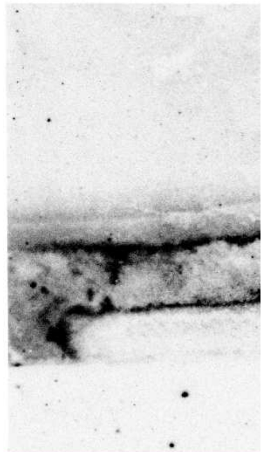


(2) Screen Deposit

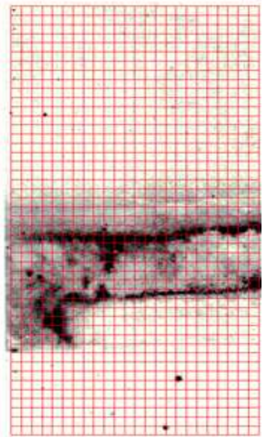
(3) Exposure Time

(4) Screen withdrawn

(5) Screen scan

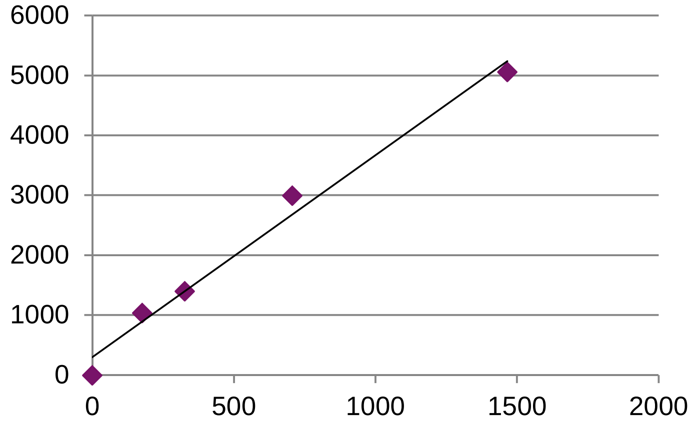


(6) Calculations



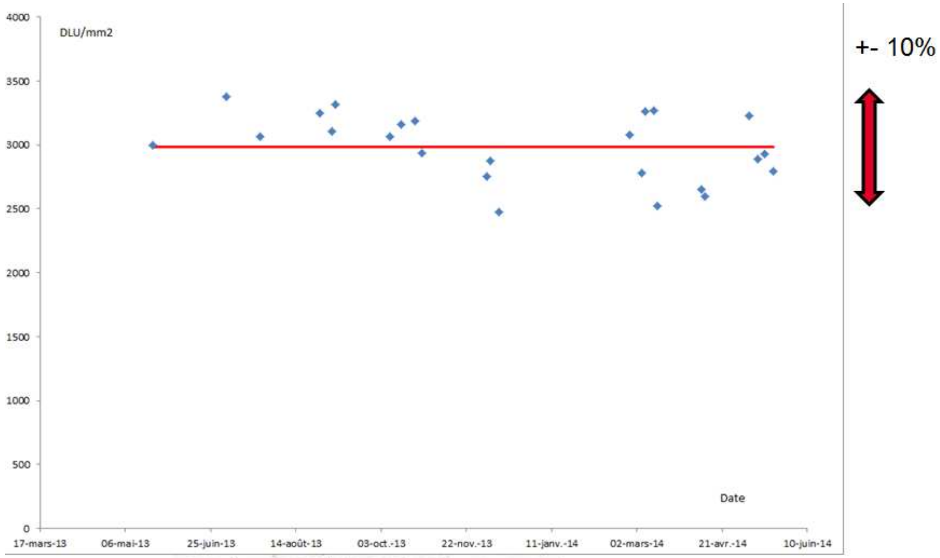
DA TECHNIQUE

- Semi quantitative values are achievable.



- Repeatability corresponds only to a few percent

Same H-3 analysis



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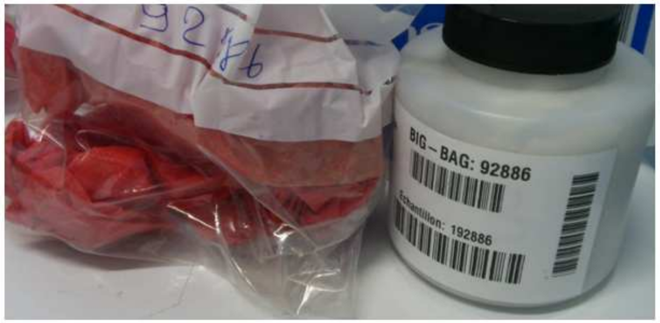
SAMPLING PROBLEMS FOR RADIOCHEMICAL ANALYSIS

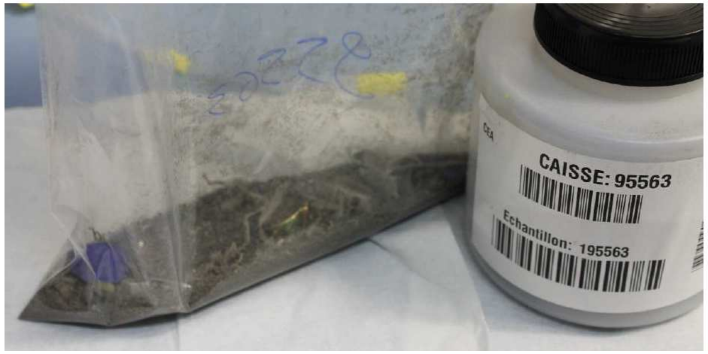
Caisse 95430

Echantillon 92886



Caisse 95434

Echantillon 95563



SAC 2 95485

How to collect 1g or even less?

DIFFERENT PROCEDURES FOR MEASUREMENTS

In Situ Sampling

Classical sampling process
by **wipe tests** =
problems for non labile radioactivity

Digestion or leaching
Alpha spectrometers
Liquid Scintillation Counting
Gamma measurements
Laboratory measurements

Container

Autoradiography

In situ measurements by Autoradiography

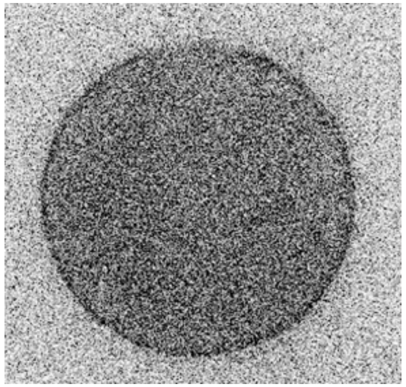
EXAMPLE OF HOME MADE STANDARDS

Preparation of homemade standards for beta analysis: CONCRETES

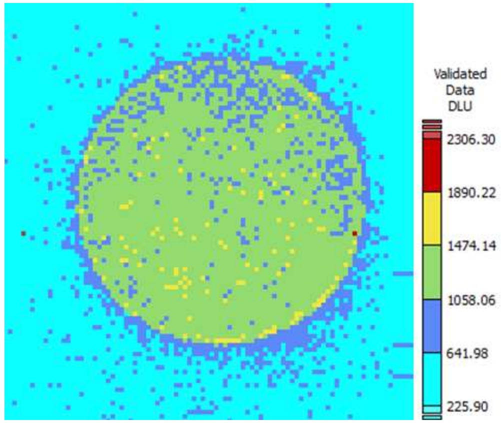
Concrete spiked at the beginning of the preparation of the cement

Can be spiked with numerous radionuclides H-3, C-14, Gamma emitters…)

Homogeneity on concrete surface (here H-3 and C-14)



Raw signal

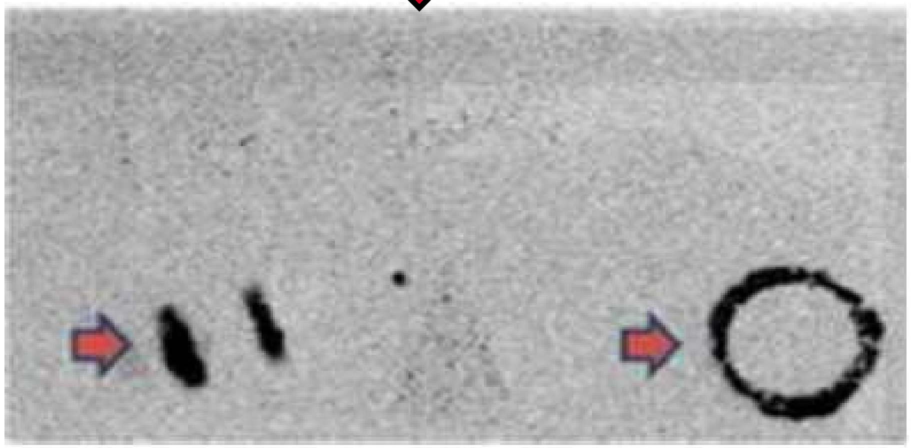


Numerical treatment to evaluate the homogeneity by digital autoradiography

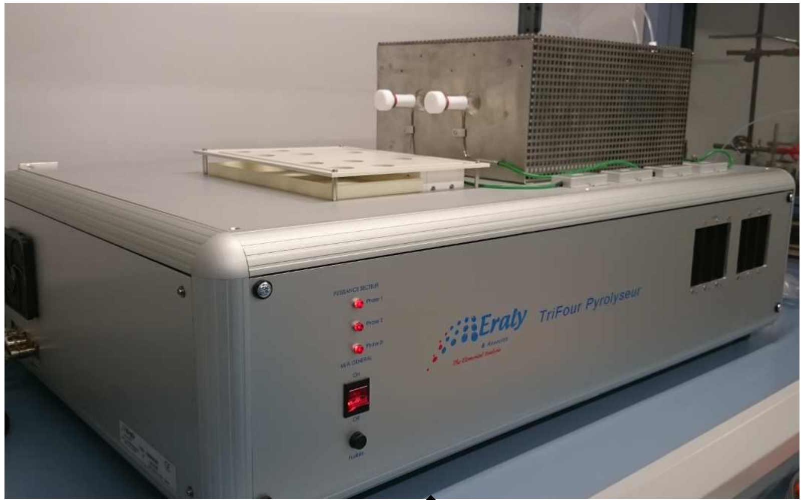
EXAMPLE OF TRITIATED WASTES

Sampling process for tritiated wastes

Autoradiography



Destructive measurement of H-3 is done by pyrolysis followed by Liquid Scintillation Counting (LSC)



Sample = 1 g

H-3 AND C-14 ANALYSIS ON WASTES

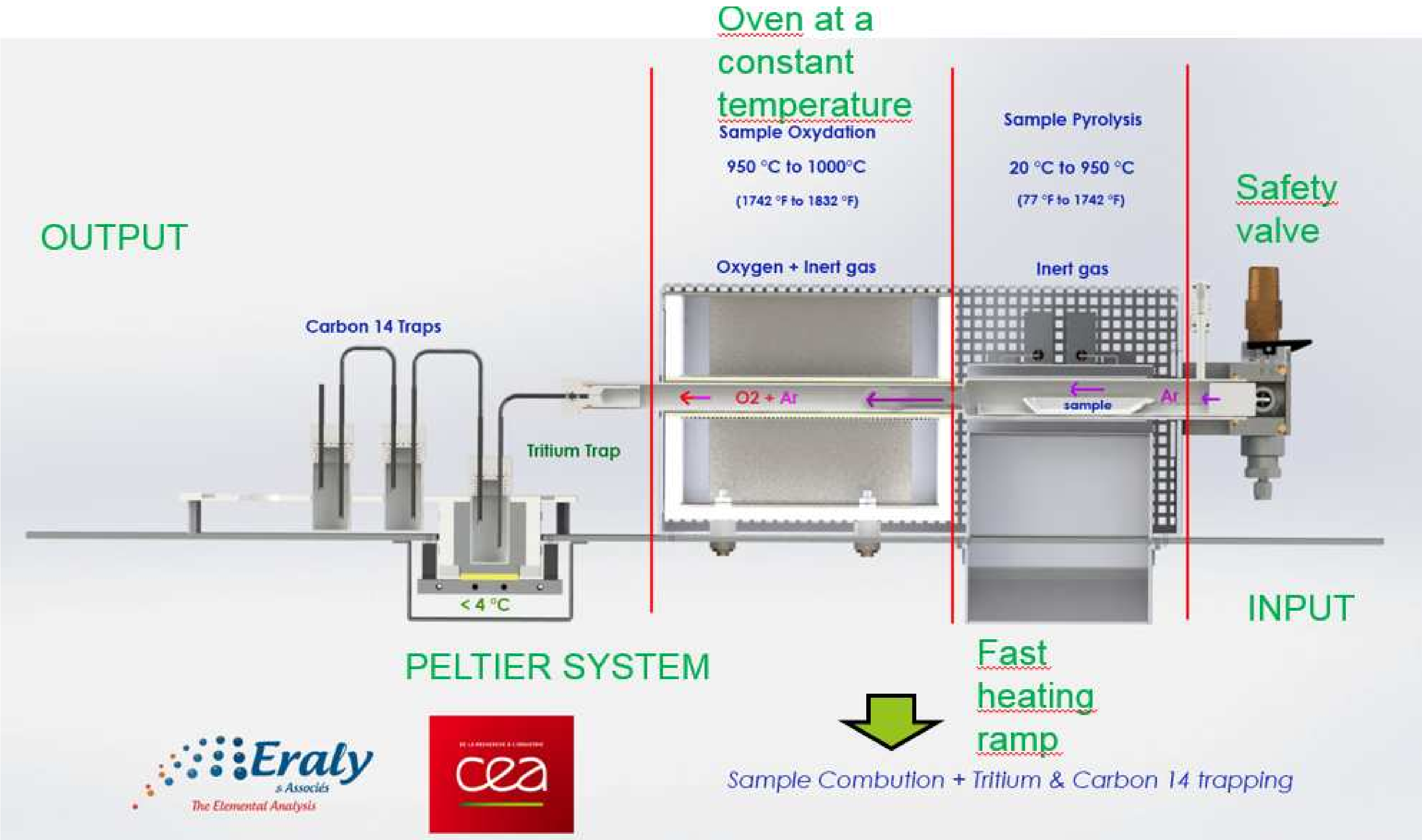
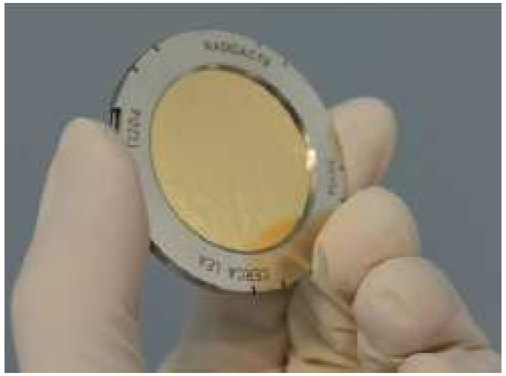
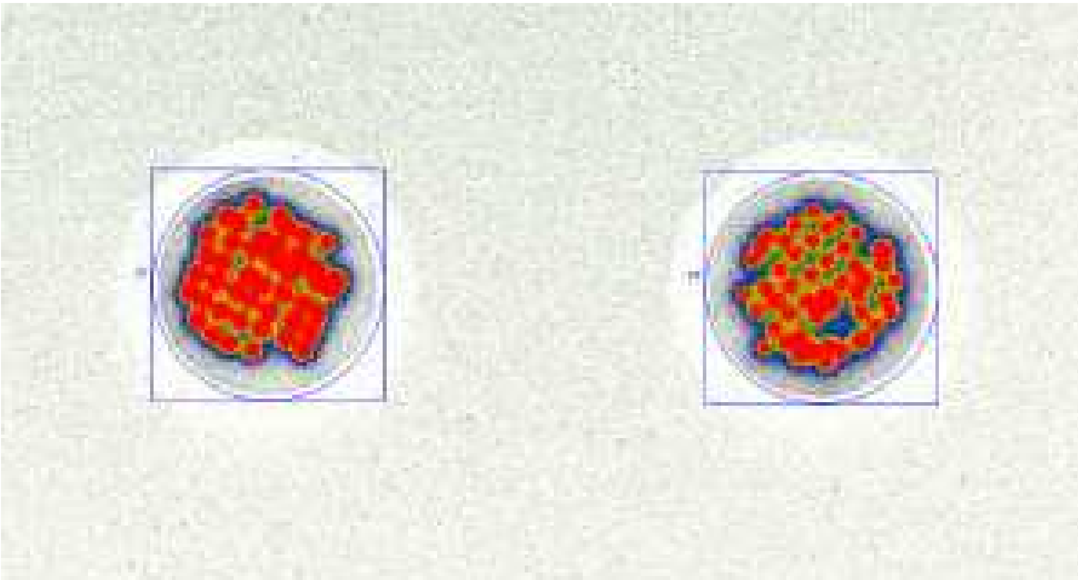


IMAGE OBTAINED WITH HIGH RESOLUTION



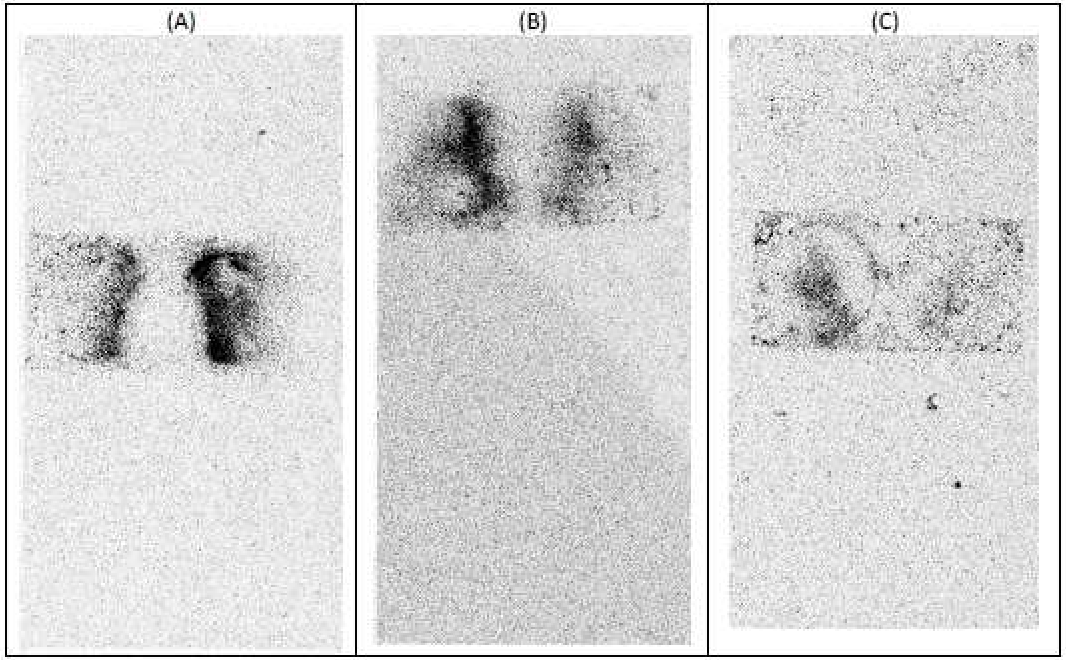


Cl-36 Sr-90

Study of sealed source by digital autoradiography – resolution used = 40 µm

EXAMPLE OF WASTES CONTAINING URANIUM

Only 3 wipes among 7 contained Uranium.

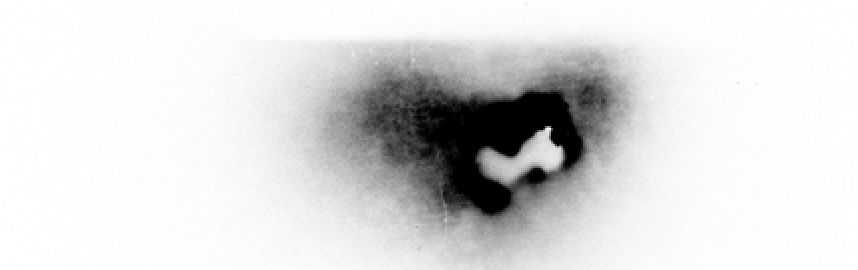


After studies with a destructive method: wipe digestion followed by alpha spectrometry and/or ICP-MS, detection limit was determined at **0.2 Bq/wipe** for Uranium (more sensitive than alpha spectroscopy).

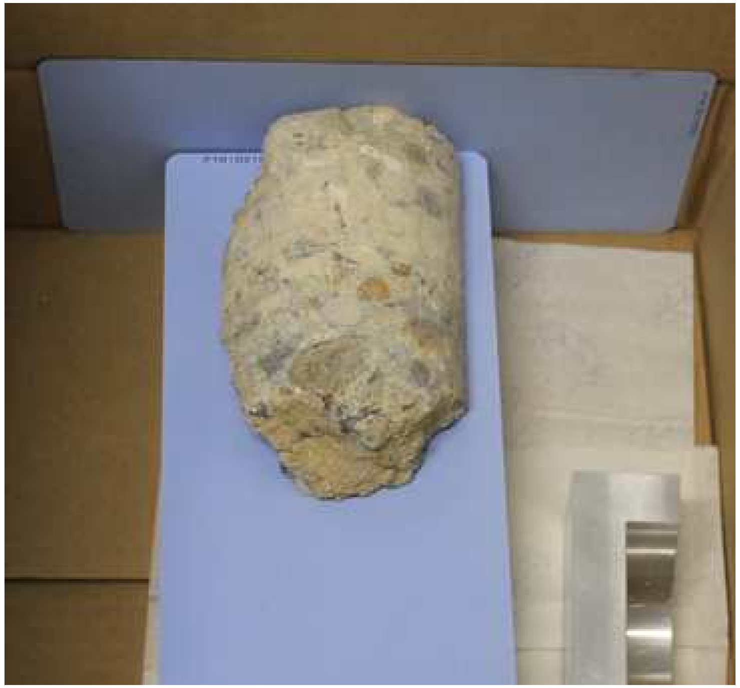
HOW TO COLLECT SAMPLES ON CORES

After surface analysis on D&D sites, determination of 3D contamination

Core made of concrete containing C-14



Activity on surface



Interesting sampling

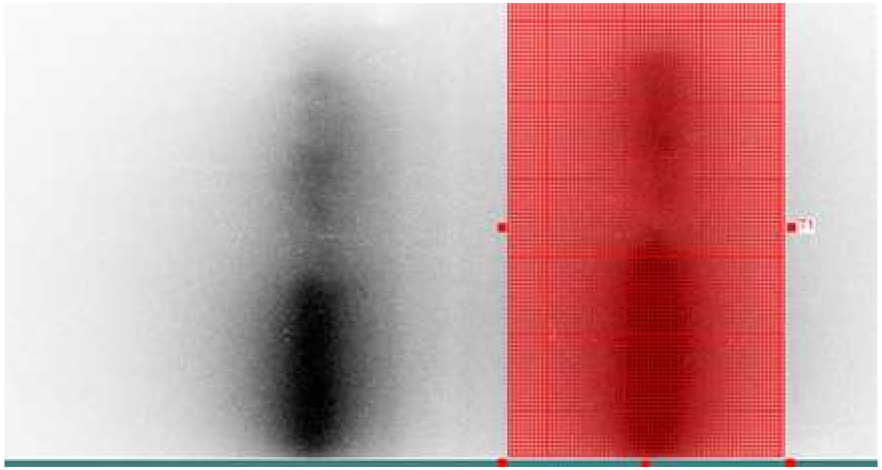
Activity in depth

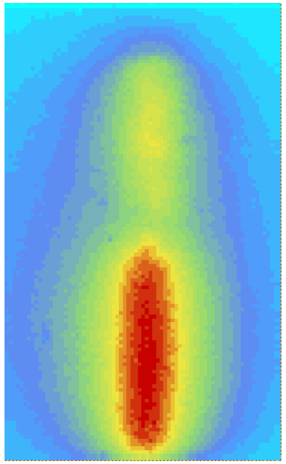
Image in real scale

HOW TO COLLECT SAMPLES ON CORES WITH HIGH AMOUNT OF RADIOACTIVITY ?

Study of alumina beads containing I-129 located inside a plastic tube

Plastic deposited on a film for **15 minutes**



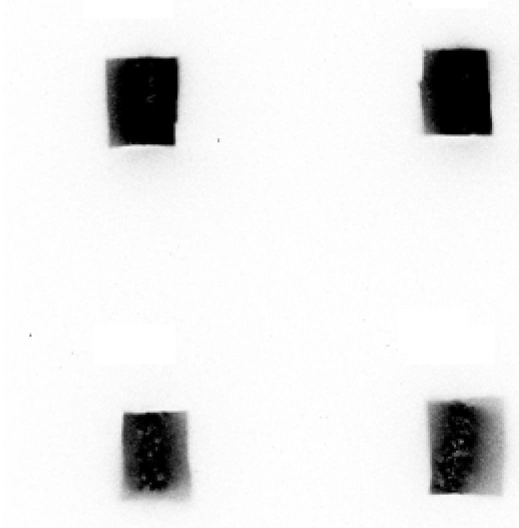


DA as efficient solution for sampling process

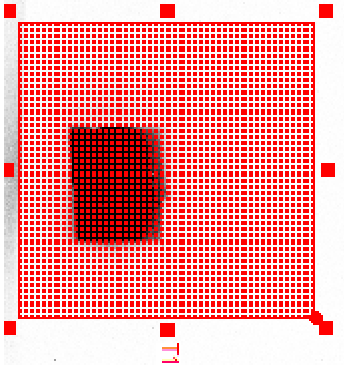
SAMPLING PROBLEM ON GRAPHITES

Graphite cores mainly containing C-14

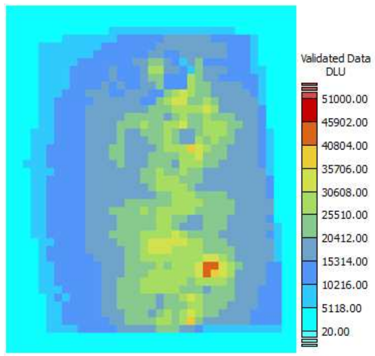
4 small cores on the same screen



Calculations



Homogeneity of the contamination at a very low scale



Digital Autoradiography is a non destructive method to assess the homogeneity

SAMPLING PROBLEM ON STEEL MATERIALS

Massive steel samples investigated by digital autoradiography

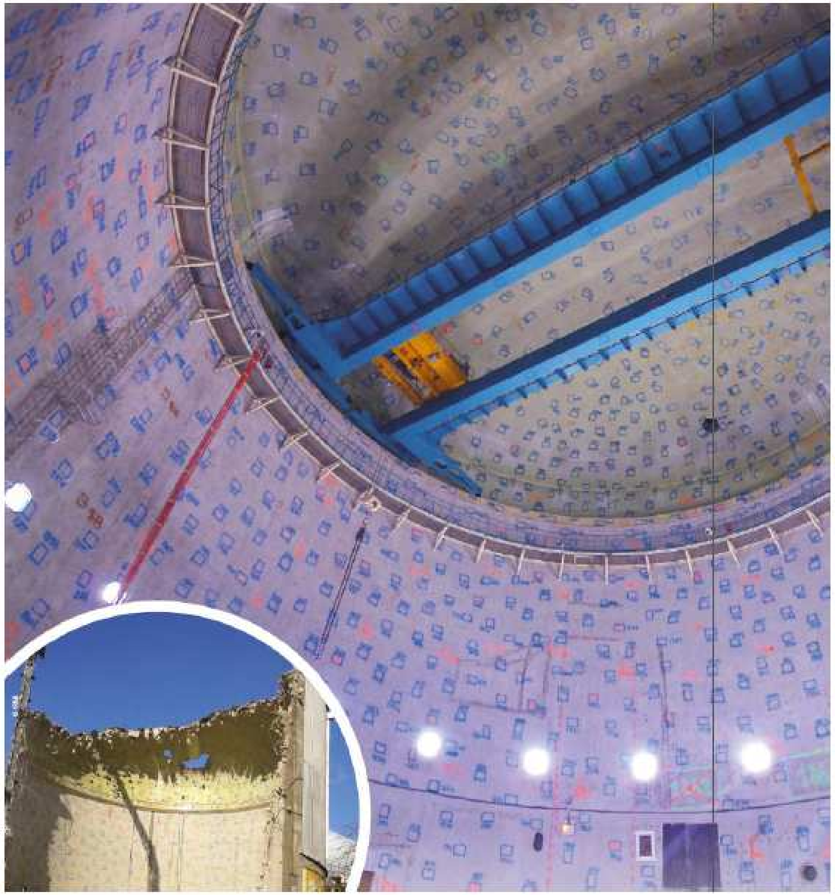
For some samples only one side is contaminated

Contamination location and homogeneity

SAMPLING FOR FINAL CONTROL TO ASSESS THE CLEARANCE LEVEL

Decommissioning requires characterization at very low level. Requirement of in situ technique to improve sampling process. 1 g sample needed for destructive analysis.

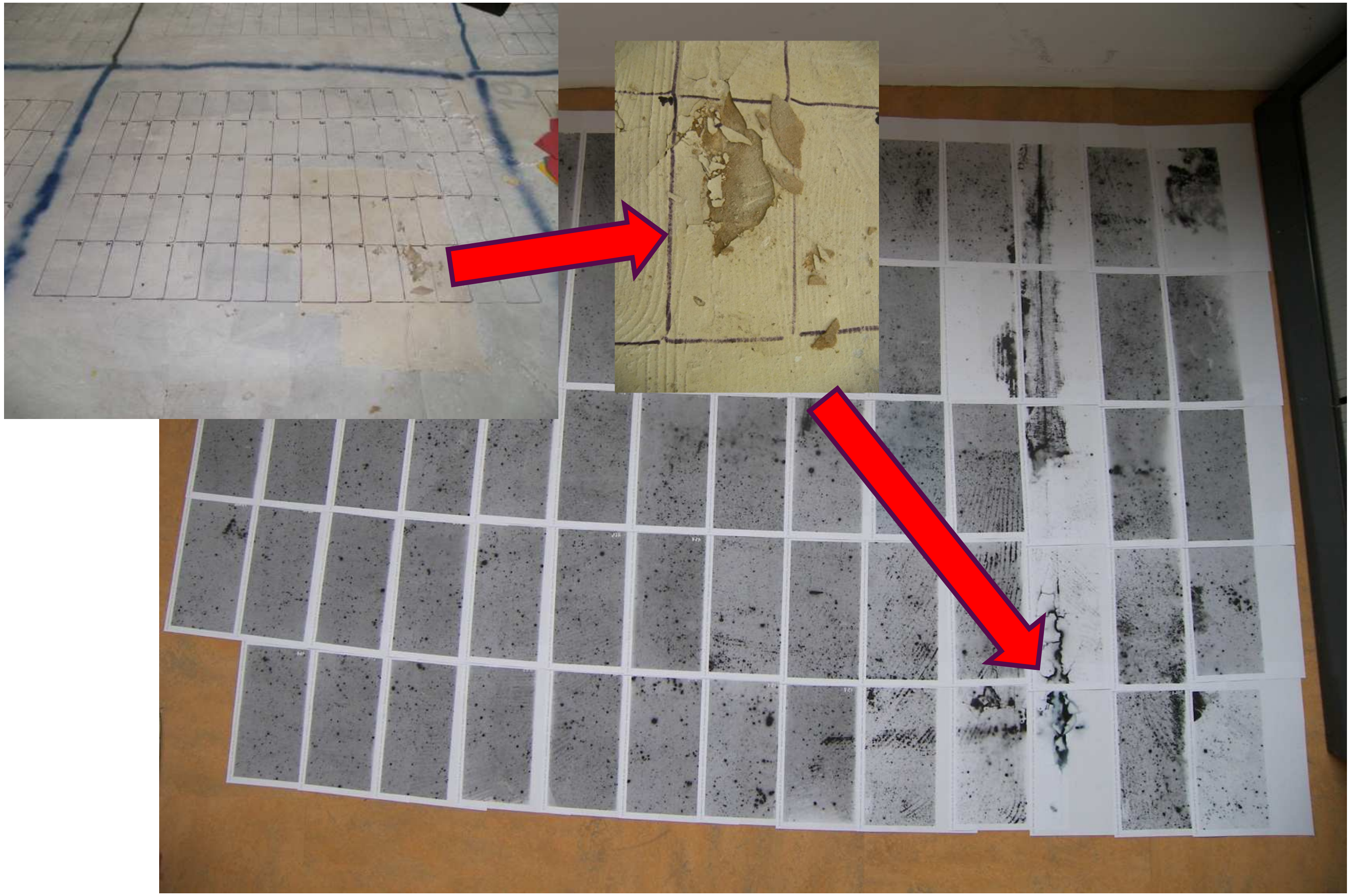
Decommissioning and dismantling



Mélusine, Grenoble

Sampling process on floor

A grid corresponding to 70 screens = 5 m^2



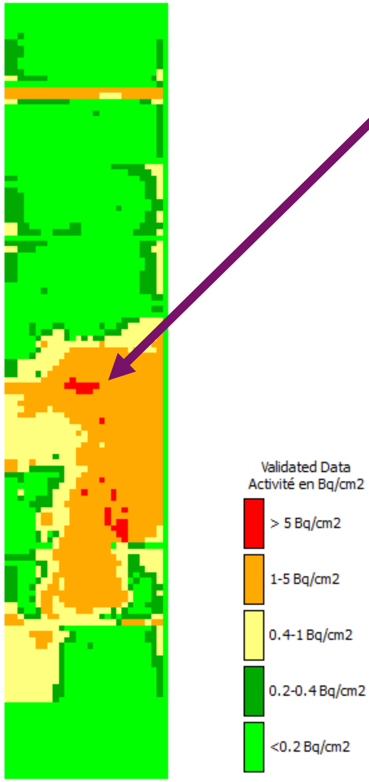
Radioactivity image, here C-14
Localisation of the sample for destructive analysis is clear

Autoradiography with raw images

OTHER EXAMPLE FOR URANIUM MEASUREMENTS

Screen on surface

1.8 m

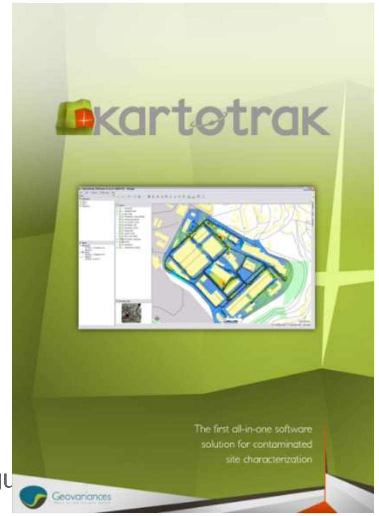


0.3 m

« Hot » spots clearly appears

Maximum activity found around 6 Bq/cm^2

Contamination shape is also an important parameter for stakeholders.

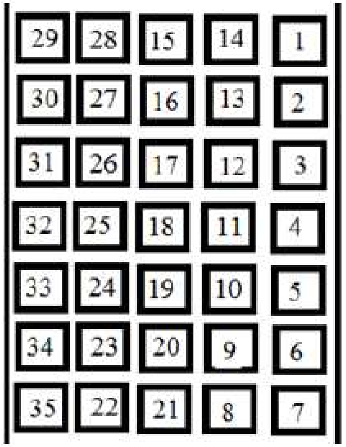


OTHER EXAMPLE FOR TRITIUM SAMPLING

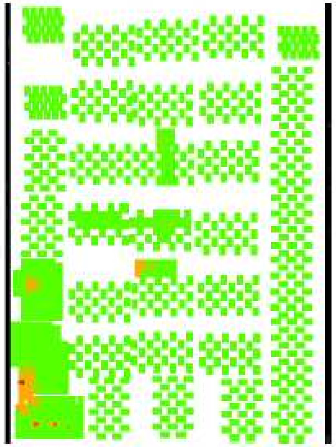
Current development: geostatistical approach

Current investigation rate:
2 weeks / 100 m²

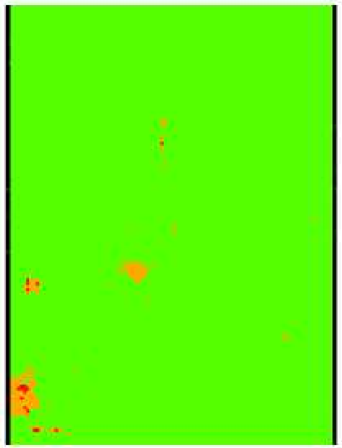
Conclusion:
Contamination accurate location
Representative and limited sampling enabling



Zoning

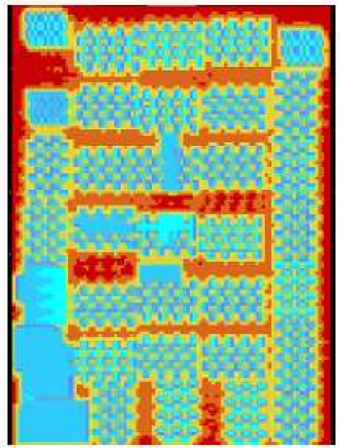


Effective mapping



Geostatistical mapping

+



Variance plot

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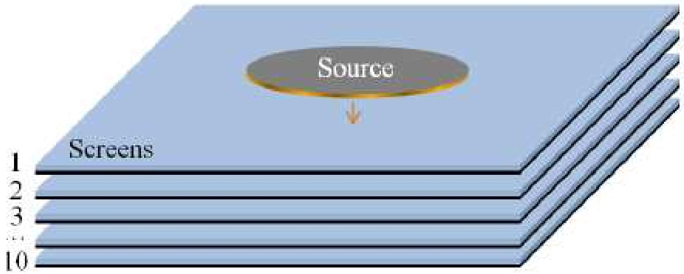
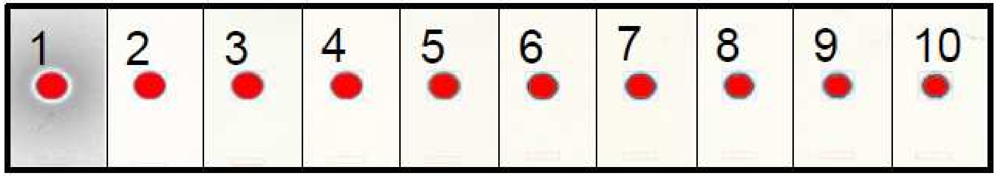
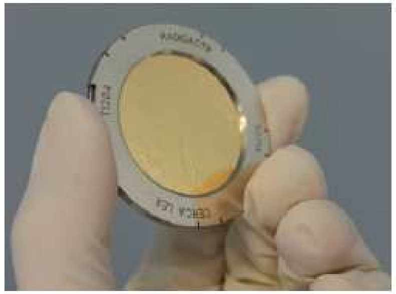
IMPROVEMENT OF SELECTIVITY

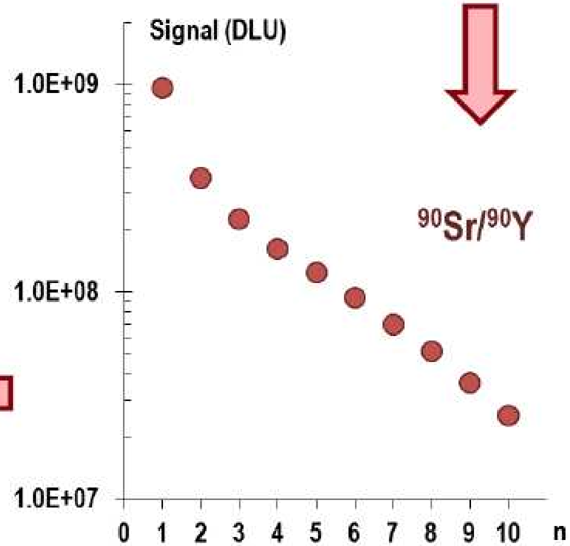
Screen stacking method (1/2)

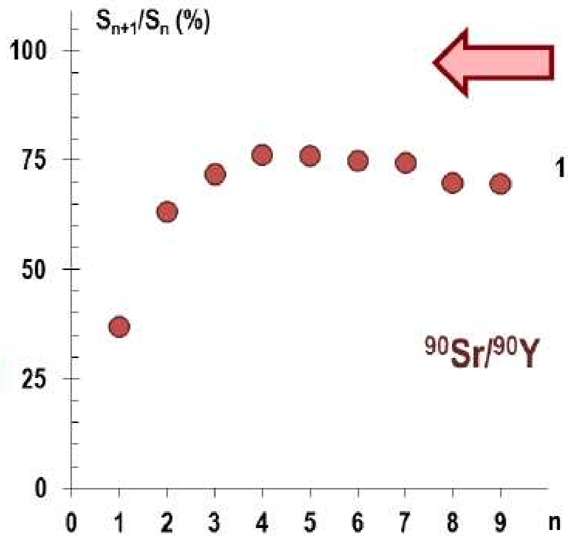


$^{90}Sr/^{90}Y$ sealed source







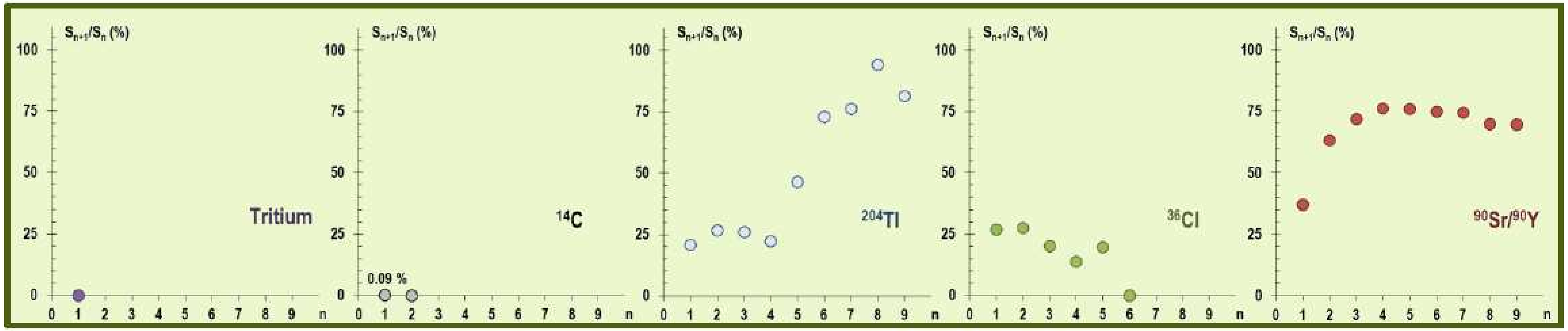


Intermediate precision

example of $^{90}Sr/^{90}Y$:
2 point sealed sources from LASE
5 sealed filtrates from LASE
1 point sealed source from INSTN
2 area sources from INSTN

S_2/S_1 = 33±6 %
S_3/S_2 = 59±6 %

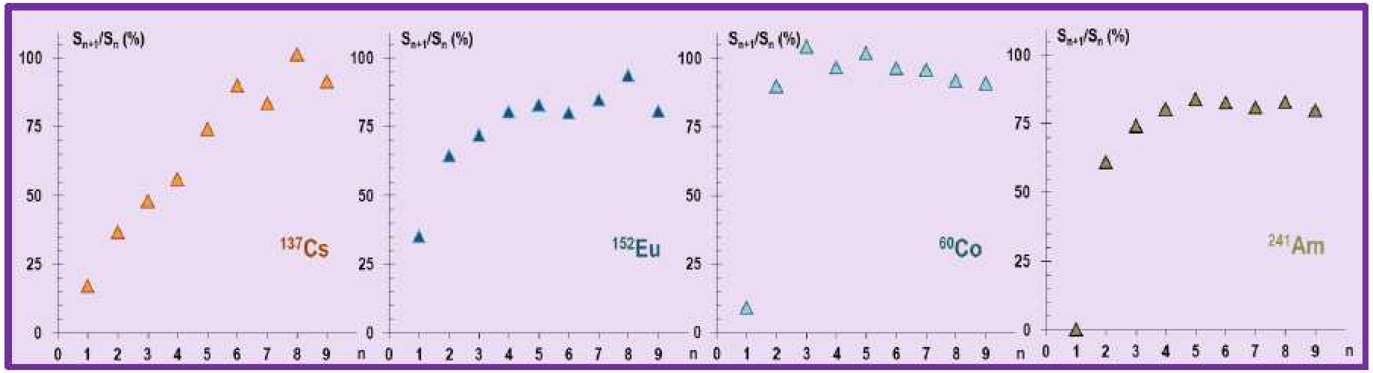
Screen stacking method (2/2)

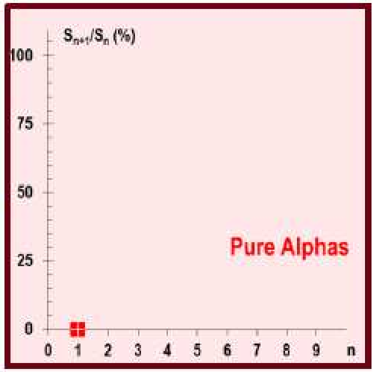


β

Radionuclides "signatures"

β+γ, α+X





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CONCLUSIONS

- Digital Autoradiography was used first because the technique is sensitive to difficult to measure radionuclides.

- Autoradiography appears to be a very interesting technique for sampling processes.

- Different developments are currently in R&D for mapping applications, to improve the sensitivity, to try to find possibilities to improve the selectivity.

- The LASE laboratory participates in numerous intercomparison tests where no sampling problem is usually encountered however it is totally different for radiochemical analysis required for radwastes matrices.

REFERENCES

References:

1) «Tritium analysis in building dismantling process using digital autoradiography » P Fichet, F Bresson, A Leskinen, F Goutelard, J Ikonen, M Siitari-Kaupi, Journal of Radioanalytical and Nuclear Chemistry, 291: 869-875 (2012).
2) « Digital autoradiography (DA) in quantification of trace level beta emitters on concrete » A Leskinen, P Fichet, M Siitari-Kauppi, Fl Goutelard, Journal of Radioanalytical and Nuclear Chemistry 298 : 153-161 (2013).
3) « A non-destructive and on-site digital autoradiography-based tool to identify contaminating radionuclide in nuclear wastes and facilities to be dismantled» R Haudebourg, P Fichet **:** J Radioanal Nucl Chem to be published 2016

Oral presentations:

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2) «Digital Autoradiography : a novel complementary technique for the investigation of radioactive contamination in nuclear facilities under dismantlement» ANNIMA 2015, Lisbon, R Haudebourg, P Fichet, F Goutelard.
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Thank you for your attention

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